@AGUPUBLICATIONS

COMMENT

Comment on "Cloud droplet spectral width relationship to CCN spectra and vertical velocity" by Hudson et al.

Yangang Liu[1], Peter H. Daum[1], and Chunsong Lu[1,2]

[1]Brookhaven National Laboratory, Upton, USA, [2]Nanjing University of Information Science and Technology, China

Correspondence to:
Y. Liu,
lyg@bnl.gov

Hudson et al. [2012, hereafter H12] have lately shown that the standard deviation (σ) of droplet size distributions was inversely related to cloud condensation nuclei (CCN) concentration at 1% supersaturation (N_{CCN}) for the in situ aircraft measurements collected during the Rain in Cumulus over the Ocean (RICO) project. Using adiabatic parcel model simulations for various values of updraft velocity (w), they further reported a tendency for the σ-N_{CCN} correlation to change signs from positive to negative as w increases beyond a certain value. The analysis of σ-N_{CCN} correlation and its dependence on w certainly add new understanding of dispersion effect (e.g., *Liu and Daum*, 2002, LD02 hereafter; *Liu et al.*, 2006b, LDY06 hereafter) and imply that like the more widely known aerosol effect on cloud droplet number concentration (N), dispersion effect may exhibit aerosol-limited and w-limited regimes such that dispersion effect can either diminish or enhance the cooling of number effect, depending on the regimes.

However, there appears to be some misunderstanding/misinterpretation of LD02 and LDY06 concerning the use of relative dispersion as a measure of spectral width. Furthermore, our examination of the data reported in Table 1 of H12 shows that there is a positive correlation between N_{CCN} and w, and thus it cannot be ruled out that the observed negative σ-N_{CCN} correlation is a manifestation of the covariation in w, or arises from the indirect effect of aerosol on cloud dynamics, or a combination of both. Below these points are detailed.

1. Measure of Spectral Width

Spectral width has been a much debated topic since late 1950s [*Squires*, 1958; *Warner*, 1969]; two commonly used measures in literature are standard deviation (σ) and relative dispersion (ε) defined as the ratio of σ to the corresponding mean radius. Note that mean diameter (MD) and the corresponding σ were used in H12. In Section 7 of H12, the authors state "… *However, most previous studies presented droplet spectral width in terms of relative dispersion ($\varepsilon = \sigma$/MD) rather than standard deviation(σ). This confused the issue of the effect of input CCN on droplet spectral width because the denominator (mean diameter) of ε is so dependent on N_{CCN} that it overwhelms the effect of N_{CCN} on σ … Liu et al.* [2006a, 2006b] *admit that much of the increase of ε attributed to higher N_{CCN} is due to the decrease of MD, but they also claim a σ increase with N_{CCN} ….*" Although we concur with most of this statement from a microphysical perspective, the authors of H12 missed the point as to why ε instead of σ has been used in our studies cited in H12.

The preference of ε over σ stems mainly from improving the parameterization of effective radius and autoconversion rate, which are essential to representing aerosol indirect effects in climate models. It has been established that effective radius (r_e) can be parameterized as [*Liu and Hallett*, 1997; *Liu and Daum*, 2000a, 2000b; LD02; *Lu and Seinfeld*, 2006]

$$r_e = \left(\frac{3}{4\pi\rho_w}\right)^{1/3} \beta \left(\frac{L}{N}\right)^{1/3},\tag{1a}$$

where ρ_w is the water density, L is the liquid water content, and the parameter β is an increasing function of ε. The difference between existing r_e parameterizations lies primarily in the specification of $\beta(\varepsilon)$, which has been shown to be well described by the expression corresponding to the Gamma droplet size distribution [*Liu and Daum*, 2000a, 2000b; *Liu et al.*, 2002]

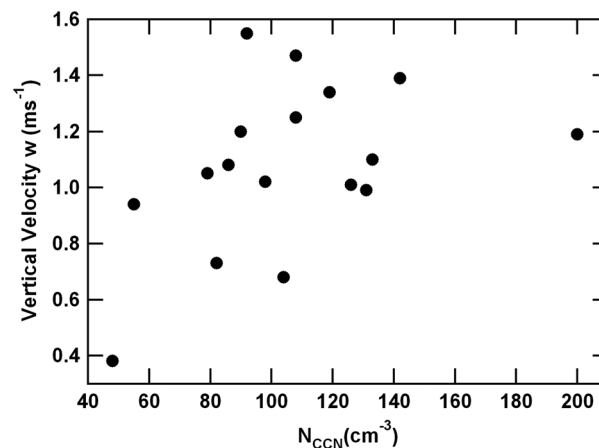$$\beta = \frac{(1 + 2\varepsilon^2)^{2/3}}{(1 + \varepsilon^2)^{1/3}}.\tag{1b}$$

Figure 1. Relationship between vertical velocity and CCN concentration at 1% supersaturation. The data are from Table 1 of H12.

The same three variables of L, N, and ε also explicitly appear in the state-of-art parameterization of autoconversion rate, which is essential for studying the second aerosol indirect effect [*Liu and Daum*, 2004; *Liu et al.*, 2006a, 2007; *Xie and Liu*, 2011; *Xie et al.*, 2013].

It is worth stressing that in bulk microphysical parameterizations for climate models, cloud properties are represented in terms of L (one-moment scheme), or L and N (two moment scheme), and further improvement demands explicit specification of ε, neither σ nor mean radius. Furthermore, although the differences between σ and ε are self-evident from their definitions, the two have often been ambiguously used in literature as a measure of spectral width, including H12, unfortunately. For example, ε was a focus of both LD02 and LDY06, and the author of H12 appeared to be aware of this according to the previous quote. However, there are at least two places of misrepresentation/confusion in H12: In P3 of H12, the authors stated, "*Negative R in Figure 1a (smaller σ for higher $N_{1\%}$) is directly opposite of the broader spectra for more polluted clouds displayed in HY97 and LD2*" Again in P8, the authors state, "*Both observations and theory indicates that droplet spectral width (σ) at low cloud altitudes in RICO shows an opposite trend with N_{CCN} from that observed and predicted by HY97 and presented by LD2 and Liu et al.* [2006b]." Although it may be inferred from their Figure 3 that the observed negative ε-$N_{1\%}$ correlation does stand in contrast with those reported in LD02 and LDY06, the quoted statements are misleading, because LD2 and LDY06 were mainly concerned with ε and its relationship to aerosols and vertical velocity, not σ, and changes in ε could be caused by changes either in σ or mean radius or both.

The P8 statement was especially misleading because H12 only presented the results of σ-N_{CCN} correlation and its variation with varying w obtained with the parcel model, not the simulated ε-N_{CCN} correlation and its variation with w. In fact, additionally showing the simulated ε-N_{CCN} relationship and its variation with w would be extremely valuable in that the result, if confirmed, could suggest the existence of w-limited and aerosol-limited regimes for dispersion effect just as for droplet concentration. *Peng et al.* [2007] reported similar results using a different parcel model.

It is worth mentioning that the preference of ε over σ in improving microphysical parameterization and quantification of aerosol indirect effects in climate models does not diminish the usefulness of σ analysis. In fact, a combined analysis of σ and ε is often more powerful to dissect microphysical mechanisms [*Pawlowska et al.*, 2006; *Pandithurai et al.*, 2012; *Prabha et al.*, 2012] and should be recommended.

2. Negative N_{CCN}-σ Correlation or a Manifestation of Correlated Dynamical Effects?

H12 reported observational evidence for a negative N_{CCN}-σ correlation and further attributed it to aerosol effects at higher w. Although both the empirical and modeling results are well based and provide some new understanding of the aerosol effect on σ, the data provided in their Table 1 suggest other possibilities as well.

Our examination of the same data shows a clear covariation of N_{CCN} and w: an increase of N_{CCN} is generally associated with an increase of w (Figure 1). Figure 2 further compares N, mean diameter, σ, and ε as a function of w. Together with the positive N_{CCN}-w correlation, the striking similarities between what is shown in Figure 2 and the corresponding plots in H12 suggest that the negative N_{CCN}-σ correlation could also be a manifestation of the effect of w covarying with N_{CCN}. Both the increase of N and the decrease of ε with increasing w are also consistent with LDY06. It is noteworthy that the contrasting changes of ε with increasing w and increasing aerosols are closely related to the supersaturation: a stronger w leads to a higher supersaturation that increases N but decreases ε whereas a higher aerosol concentration leads to a higher N but lower supersaturation. Also noteworthy is that the negative correlations between ε (σ) and w arise largely from the data points
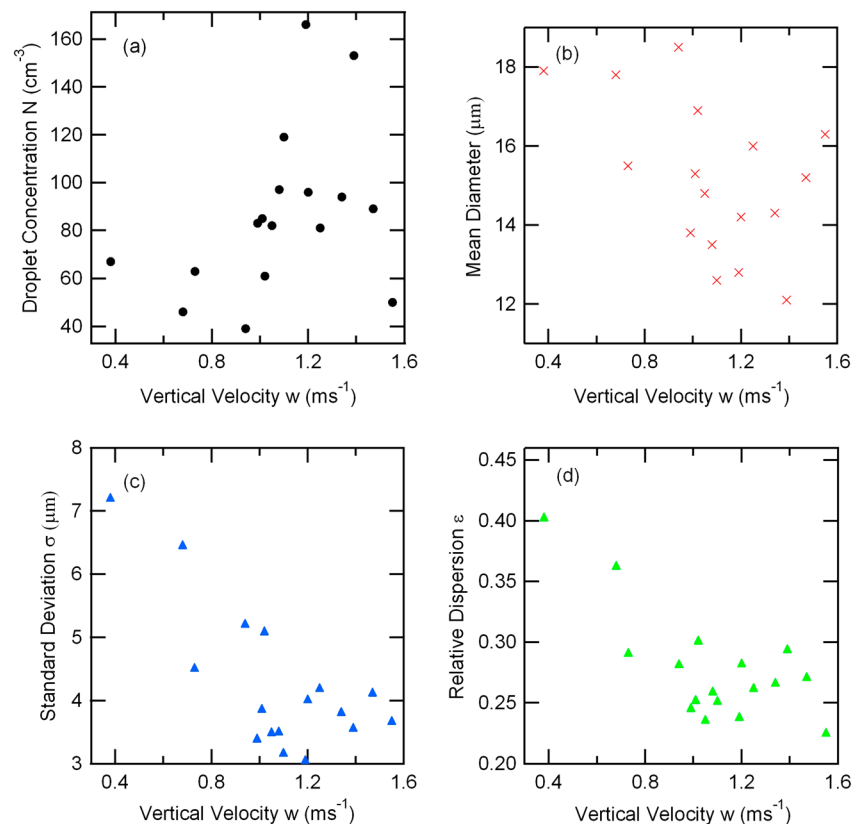
Figure 2. Observational relationships of vertical velocity to (a) droplet concentration, (b) mean diameter, (c) standard deviation, and (d) relative dispersion. The data are from Table 1 of H12.

with lower w ($< 1\,ms^{-1}$); for higher w the decreasing trend is not obvious. This result could suggest that the effect of w on ε (σ) starts to level off after w reaches a certain value, or the value of ε (σ) is close to the limit of the probe used, or is due to poor statistics; more studies are needed on this front.

Although the covariation of N_{CCN} and w does not necessarily mean a cause-and-effect relationship, one cannot completely rule out an even more interesting possibility that the negative N_{CCN}-σ correlation results from the indirect effect of aerosol on vertical velocity according to some modeling studies [*Xue and Feingold*, 2006; *Lu and Seinfeld*, 2006]. These modeling studies suggested that increasing aerosols tend to strengthen vertical motion and entrainment processes in clouds by altering condensation and evaporation rates and latent heating. The potential aerosol effect on entrainment processes adds an additional layer of difficulty to the already vexing problem of untangling aerosol effects from dynamical effects. A positive correlation between N_{CCN} and w was also reported in *Berg et al.* [2011]. Note that the CO concentration was used as a proxy of aerosol loading. Being coincidence or aerosol dynamical effects, the association between the subcloud N_{CCN} and in-cloud w makes the data interpretation more challenging and calls for further investigation into the separation of aerosol effects from dynamical effects. An examination of w and aerosol properties below clouds would also help resolve the issue: a coexisting positive w-N_{CCN} correlation just below clouds suggests a common mechanism for the increases in w and N_{CCN}.

References

Berg, L. K., C. M. Berkowtiz, J. C. Barnard, G. Senum, and S. R. Springston (2011), Observations of the first aerosol indirect effect in shallow cumuli, *Geophys. Res. Lett.*, *38*, L03809, doi:10.1029/2010GL046047.

Hudson, J. G., S. Noble, and V. Jha (2012), Cloud droplet spectral width relationship to CCN spectra and vertical velocity, *J. Geophys. Res.*, *117*, D11211, doi:10.1029/2012JD017546.

Liu, Y., and J. Hallett (1997), The "1/3" power-law between effective radius and liquid water content, *Q. J. R. Meteorol. Soc.*, *123*, 1789–1795.

Liu, Y., and P. H. Daum (2000a), Spectral dispersion of cloud droplet size distributions and the parameterization of cloud droplet effective radius, *Geophys. Res. Lett.*, *27*, 1903–1906.

Liu, Y., and P. H. Daum (2000b), Which size distribution function to use for studies related effective radius. Proceeding of the 13th International Conf. On Clouds and Precipitation, 586-589, Reno, USA, 14-18 August 2000.

Liu, Y., and P. H. Daum (2002), Indirect warming effect from dispersion forcing, *Nature*, *419*, 580–581.

Liu, Y., and P. H. Daum (2004), Parameterization of the autoconversion process, Part I: Analytical formulation of the Kessler-type parameterizations, *J. Atmos. Sci.*, *61*, 1539–1548.

Liu, Y., P. H. Daum, S. K. Chai, and F. Liu (2002), Cloud parameterizations, cloud physics, and their connections: An overview, *Recent Res. Dev. Geophys.*, *4*, 119–142.

Liu, Y., P. H. Daum, R. McGraw, and M. Miller (2006a), Generalized threshold function accounting for effect of relative dispersion on threshold behavior of autoconversion process, *Geophys. Res. Lett.*, *33*, L11804, doi:10.1029/2005GL0255000.

Liu, Y., P. H. Daum, and S. Yum (2006b), Analytical expression for the relative dispersion of the cloud droplet size distribution, *Geophys. Res. Lett.*, *33*, L02810, doi:10.1029/2005GL024052.

Liu, Y., P. H. Daum, R. McGraw, M. Miller, and S. Niu (2007), Theoretical expression for the autoconversion rate of droplet concentration, *Geophys. Res. Lett.*, *34*, L16821, doi:10.1029/2007GL030389.

Lu, M.-L., and J. H. Seinfeld (2006), Effect of aerosol number concentration on cloud droplet dispersion: A large-eddy simulation study and implications for aerosol indirect forcing, *J. Geophys. Res.*, *111*, D02207, doi:10.1029/2005JD006419.

Pandithurai, G., S. Dipu, T. V. Prabha, R. S. Maheskumar, J. R. Kulkarni, and B. N. Goswami (2012), Aerosol effect on droplet spectral dispersion in warm continental cumuli, *J. Geophys. Res.*, *117*, D16202, doi:10.1029/2011JD016532.

Pawlowska, H., W. W. Grabowski, and J. L. Brenguier (2006), Observations of the width of cloud droplet spectra in stratocumulus, *Geophys. Res. Lett.*, *33*, L19810, doi:10.1029/2006GL026841.

Peng, Y., U. Lohmann, R. Leaitch, and M. Kulmala (2007), An investigation into the aerosol dispersion effect through the activation process in marine stratus clouds, *J. Geophys. Res.*, *112*, D1117, doi:10.1029/2006JD007401.

Prabha, T. V., S. Patade, G. Pandithurai, A. Khain, D. Axisa, P. Pradeep-Kumar, R. S. Maheshkumar, J. R. Kulkarni, and B. N. Goswami (2012), Spectral width of premonsoon and monsoon clouds over Indo-Gangetic valley, *J. Geophys. Res.*, *117*, D20205, doi:10.1029/2011JD016837.

Squires, P. (1958), The microstructure and colloidal stability of warm clouds. Part I: The relation between structure and stability, *Tellus*, *10*, 256–261.

Warner, J. (1969), The microstructure of cumulus cloud: Part I. General features of the droplet spectrum, *J. Atmos. Sci.*, *26*, 1049–1059.

Xie, X., and X. Liu (2011), Effects of spectral dispersion on clouds and precipitation in mesoscale convective systems, *J. Geophys. Res.*, *116*, D06202, doi:10.1029/2010JD014598.

Xie, X., X. Liu, Y. Peng, Y. Wang, Z. Yue, and X. Li (2013), Numerical simulation of clouds and precipitation depending on the relationship between aerosol and cloud droplet relative dispersion, *Tellus*, *B65*, 19,054.

Xue, H., and G. Feingold (2006), Large-eddy simulations of trade wind cumuli: Investigation of aerosol indirect effects, *J. Atmos. Sci.*, *63*(6), 1605–1622, doi:10.1175/jas3706.1.